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14. ABSTRACT The completed research aimed to introduce the Optimal Dynamic Detection of Explosives (ODD-Ex) as a means to harness the best capabilities of advanced laser technology to significantly enhance the standoff detection of explosives. The core of the ODD-Ex technique rested on employing optimally shaped laser pulses to simultaneously enhance sensitivity to explosive signatures while reducing the influence of noise and signals from background interferents in the field. These goals were addressed by operating in an optimal non-linear fashion, with a single shaped pulse inherently containing within it coherently locked control and probe sub-pulses. The research teams used this technique to intrinsically provide orthogonal broad spectral information for data fusion, all from a single optimal pulse. The research program entailed (i) theoretical studies of the basic ODD-Ex concepts, (ii) modeling and simulations to assess the best operational procedures and (iii) laboratory demonstrations of the key detection principles.					
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Final Technical Report

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PI Name: Herschel Rabitz

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Final Technical Report for May 1, 2008 to July 31, 2011:

This is a final report on research conducted to explore the ability to optimally utilize laser resources for the remote detection of explosives. The research performed under this grant encompassed two main focuses: (1) investigating the use of laser control resources to manage the competition between selectivity and sensitivity in the detection of explosive agents and (2) testing various methods of utilizing ultrafast broad bandwidth pulse shaping for explosive agent detection.

(1) List of Papers Published Under ONR Sponsorship:

1. Multiobjective adaptive feedback control of two-photon absorption coupled with propagation through a dispersive medium, F. Laforge, J. Roslund, O. Shir, H. Rabitz, *Phys. Rev. A*, **84**, 013401 (2011).
2. Control of nitromethane photoionization efficiency with shaped femtosecond pulses, J. Roslund, O. Shir, A. Dogariu, R. Miles, H. Rabitz, *J. Chem. Phys.*, **134**, 154301 (2011).
3. Resolution of strongly competitive product channels with optimal dynamic discrimination: Application to flavins, J. Roslund, M. Roth, L. Guyon, V. Boutou, F. Courvoisier, J.-P. Wolf, H. Rabitz, *J. Chem. Phys.*, **134**, 034511 (2011).
4. Optical Dynamic Detection of Explosives, Moore, D.S., Rabitz, H., McGrane, S.D., Greenfield, M., et. al., *SPIE*, **7304**, 730413 (2011).

(2) Scientific Personnel Supported by this Project:

Princeton University

Herschel Rabitz, Principal Investigator

Francois Laforge, Postdoc

Jon Rosland, Graduate Student

Los Alamos National Laboratory

David S. Moore, Co-Principal Investigator

(3) **Report of Inventions:** None.

(4) **Scientific Progress and Accomplishments:**

1. Multiobjective adaptive feedback control of two-photon absorption coupled with propagation through a dispersive medium, F. Laforge, J. Roslund, O. Shir, H. Rabitz, *Phys. Rev. A*, **84**, 013401 (2011).

This research used shaped femtosecond laser pulses to control the two-photon absorption (TPA) of coumarin 153 in a dispersive toluene medium. The dispersive medium reshapes the pulse along the optical path, and management of this effect was used to achieve spatial localization of TPA. Other control objectives were successfully implemented, including dual localization and high resolution local optimization of TPA. The solutions to these objectives were explored by means of evolutionary single- and multi-objective algorithms within a laboratory feedback loop.

2. Control of nitromethane photoionization efficiency with shaped femtosecond pulses, J. Roslund, O. Shir, A. Dogariu, R. Miles, H. Rabitz, *J. Chem. Phys.*, **134**, 154301 (2011).

The applicability of adaptive femtosecond pulse shaping was studied for achieving selectivity in the photoionization of low-density polyatomic targets. In particular, optimal dynamic discrimination (ODD) techniques exploited intermediate molecular electronic resonances that allow a significant increase in the photoionization efficiency of nitromethane with shaped near-infrared femtosecond pulses. The intensity bias typical of high-photon number, nonresonant ionization is accounted for by reference to a strictly intensity-dependent process. Closed-loop adaptive learning was then able to discover a pulse form that increased the ionization efficiency of nitromethane by approximately 150%. The optimally-induced molecular dynamics resulted from entry into a region of parameter space inaccessible with intensity-only control. Finally, the discovered pulse shape was demonstrated to interact with the molecular system in a coherent fashion as assessed from the asymmetry between the response to the optimal field and its time-reversed counterpart.

3. Resolution of strongly competitive product channels with optimal dynamic discrimination: Application to flavins, J. Roslund, M. Roth, L. Guyon, V. Boutou, F. Courvoisier, J.-P. Wolf, H. Rabitz, *J. Chem. Phys.*, **134**, 034511 (2011).

Fundamental molecular selectivity limits were probed by exploiting laser-controlled quantum interferences for the creation of distinct spectral signatures in two flavin molecules, erstwhile nearly indistinguishable via steady-state methods. Optimal dynamic discrimination (ODD) used optimally shaped laser fields to transiently amplify minute molecular variations that would otherwise go unnoticed with linear absorption and fluorescence techniques. ODD is experimentally demonstrated by combining an optimally shaped UV pump pulse with a time-delayed, fluorescence-depleting IR pulse for discrimination amongst riboflavin and flavin mononucleotide in aqueous solution, which are structurally and spectroscopically very similar. Closed-loop, adaptive pulse shaping discovered a set of UV pulses that induced disparate responses from the two flavins and allowed for concomitant flavin discrimination of similar to 16 sigma. Additionally, attainment of ODD permitted quantitative, analytical detection of the

individual constituents in a flavin mixture. The successful implementation of ODD on quantum systems of such high complexity bodes well for the future development of the field and the use of ODD techniques in a variety of demanding practical applications.

4. Optical Dynamic Detection of Explosives, Moore, D.S., Rabitz, H., McGrane, S.D., Greenfield, M., et. al., *SPIE*, **7304**, 730413 (2011).

In this work, the Los Alamos team focused on applying the ODD-Ex paradigm to coherent anti-Stokes Raman spectroscopy (CARS) since it has already proven to be a very good method for detecting and identifying explosive materials. Hence, ODD-Ex improved on an already-existing laser-based spectroscopy technique. The Los Alamos team has shown that the explosive signal can be maintained while reducing the signal from interfering materials present by several orders of magnitude, and the team expects even broader application will be possible with the next generation of studies currently underway.

In this work, the Princeton team focused on exploring the fundamental chemical-selectivity limits of ODD and extending its general applicability for domains that include atmospheric investigation, biological discrimination, and rapid molecular interrogation. One approach undertaken by the Princeton team utilized shaped femtosecond pulses to selectively ionize a target material in the vapor phase while minimizing indiscriminate or deleterious ionization of background interferents. The resultant laser-induced plasma then scatters high-frequency microwaves that are coherently detected with established homodyne techniques for excellent long-range detection possibilities. This unique combination of specificity afforded through optical pulse shaping and high detection sensitivity from radar scattering has yielded dramatic improvements in ionization selectivity compared to unshaped pulses. Further advances are expected with continued experimentation.

5. **Technology Transfer:** None.